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Nonlinearity parameter, *B/A*, of pure liquids and binary liquid mixtures at elevated pressures

V K Singh
Department of Physics, VSSD (PG) College, Kanpur-208 002, Uttar Pradesh, India
E-mail: singhvk_singhvk@yahoo.co.in

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Abstract : Nonlinearity parameter (*B/A*) has been evaluated for three pure liquids, namely : toluene, o-xylene, aniline and two binary mixtures, namely : toluene + o-xylene, toluene + aniline at elevated pressures at 303.15 K with the help of four different methods using the experimental data of ultrasonic velocity and density. A comparative study of the results obtained from different methods has been carried out in order to review different approaches in the light of molecular structure and intermolecular interactions.

Keywords : Nonlinearity parameter, binary mixtures at elevated pressures, ultrasonic velocity.

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1. Introduction

It has become of much interest to predict the extent of various kinds of nonlinear effects, which are due to deviation from linearity in ordinary acoustics [1–5]. The nonlinearity parameter *B/A* is a basic parameter for determining the degree of waveform distortion. From the knowledge of this parameter, one can gain information about some physical properties [6,7] of the liquids such as internal pressure, intermolecular spacing, acoustic scattering and structural behaviour *etc*.

Study of nonlinearity parameter of liquid mixtures is important in view of the information it may yield on the interaction in liquid mixtures. *B/A* values of the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquid [8] which may be considered to be true for the liquid mixtures as well.

Nonlinearity parameter plays a significant role in nonlinear acoustics and its determination is of increasing interest in a number of areas ranging from underwater acoustics to medical science. Also, measurement of this parameter for biological media [9–11] including living tissues is one of the more recent interests as it can provide important information for the ultrasonic application in biological research related to diagnosis and therapy. During recent years [12–21], a number of experimental and theoretical studies have been performed on the nonlinearity parameter of liquids and liquid mixtures but to the best of our knowledge, computation of *B/A* at elevated pressures has not been done so far.

The present work deals with the computation of acoustic nonlinearity parameter *B/A* of three pure liquids and two binary liquid mixtures at elevated pressures and at temperature of 303.15 K using four different approaches. The different methods used are Ballou's empirical relation, Hartmann's relation, Tong-Dong method using Schaaffs equation for sound velocity and Sharma's method. Furthermore, a comparative study of *B/A* values obtained from different methods has also been carried out in order to review above mentioned approaches and the merits and demerits of these different methods are discussed in the light of molecular structure and intermolecular interactions.

2. Theoretical

According to the empirical rule of Ballou employed by Hartmann [22], there is a linear relation between the nonlinearity parameter of liquids and reciprocal sound speed as

$$\frac{B}{A} = -0.5 + \frac{(1.2\times10^4)}{u}, \tag{1}$$

where u is the sound velocity expressed in ms^{-1}.

Hartmann [22] has shown theoretically, the physical basis for eq. (1) assuming that the intermolecular potential energy is the dominant factor in determining sound speed, and its derivatives in liquids. The expression for nonlinearity parameter due to Hartmann [22] is

$$\frac{B}{A} = 2 + \frac{(0.98\times10^4)}{u} \tag{2}$$

Nonlinearity parameter B/A can also be calculated from the modified thermodynamic equation of state [3] for liquids, namely,

$$P = P_o + A\left(\frac{\rho-\rho_o}{\rho_o}\right) + \frac{B}{2}\left(\frac{\rho-\rho_o}{\rho_o}\right)^2, \tag{3}$$

where

$$A = \rho_o\left(\frac{\partial P}{\partial \rho}\right)_S\Bigg|_{\rho=\rho_o} \quad\text{and}\quad B = \rho_o^2\left(\frac{\partial^2 P}{\partial \rho^2}\right)_S\Bigg|_{\rho=\rho_o}. \tag{4}$$

Since $u_o^2 = \left(\frac{\partial P}{\partial \rho}\right)_S\Big|_{\rho=\rho_o}$, we get

$$\frac{B}{A} = \rho\left(\frac{\partial u^2}{\partial P}\right). \tag{5}$$

Here, the subscript 'o' is omitted for brevity.

Tong *et al* [23] applied Schaaffs equation [24] for sound velocity in eq. (5) and obtained an equation for B/A as

$$\frac{B}{A} = J(o) + J(x), \tag{6}$$

where $$J(o) = \left(1-\frac{1}{\gamma}\right)\frac{u^2\rho\beta_T}{\alpha T} = \frac{\gamma-1}{\alpha T} \tag{7}$$

and $$J(x) = \frac{2(3-2x)^2}{3(x-1)(6-5x)}. \tag{8}$$

Here, γ is the ratio of specific heats, ρ the density, α the thermal expansivity, β_T the isothermal compressibility and x is the real volume of a mole of molecules given by

$$x = \frac{V}{b} = \frac{M}{\rho b},$$

where V is the molar volume and b is the van der Waals constant. Here, we have the condition [23] for x as $1 < x < 1.2$.

From eq. [8], it is evident that as the value of x tends to 1.2, the value of $J(x)$ tends to infinity which is an absurd result. Hence, value of x plays a very important role in determining B/A values using Tong-Dong method.

General expression for the nonlinearity parameter in terms of the acoustical parameters of liquids, has been derived using the expression for the sound velocity u, and introducing the contribution due to isochoric K'', and isobaric K, acoustical parameters. The properties needed have been deduced with the help of thermal expansion coefficient α of the liquid. Beyer's parameter of nonlinearity due to Sharma [19] method, may be expressed as

$$\frac{B}{A} = 2K + 2\gamma K'', \tag{9}$$

where γ is the heat capacities ratio.

3. Results and discussion

The values of the nonlinearity parameter B/A of three pure liquids and two binary liquid mixtures, have been calculated at elevated pressures ranging from 0.1 MPa to 160 MPa and at temperature of 303.15 K with the help of four different methods : Ballou's empirical relation [eq. (1)], Hartmann's theoretically derived relation [eq. (2)], Tong-Dong method using Schaaffs equation for sound velocity [eq. (6)], and Sharma's method [eq. (9)].

Under present investigation, the pure liquids are toluene, o-xylene and aniline, and the binary liquid mixtures are toluene + o-xylene and toluene + aniline. The different parameters of these liquids and liquid mixtures needed for the computation, have been obtained with the help of well-known thermodynamic relations and recently proposed empirical relations [25,26]. Thermal expansion coefficient α and isothermal compressibility β_T are obtained using the relations [25,26] :

$$\alpha = \frac{75.6\times10^{-3}}{T^{1/9}u^{1/2}\rho^{1/3}} \tag{10}$$

and

$$\beta_T = \frac{17.1\times10^{-4}}{T^{4/9}\rho^{4/3}u^2} \tag{11}$$

where u and ρ are ultrasonic velocity and density at temperature T. The heat capacities ratio γ is calculated from the well-known thermodynamic relation

$$= \frac{\beta_T}{\beta_S}, \tag{12}$$

where isentropic compressibility β_S is obtained as

$$\beta_S = \frac{1}{u^2 \rho}. \tag{13}$$

Experimental values of ultrasonic velocity and density are taken from the literature [27]. The calculated values of nonlinearity parameter B/A for pure liquids and binary liquid mixtures using different methods are reported in different tables.

A close perusal of Tables 1–3 reveals that the values of B/A calculated using eqs. (1) and (2) show a decreasing trend with increase of pressure, whereas, the B/A values obtained from eq. (9) increase with increasing pressure. However, the variation in the B/A values due to this method is very small. Table 1 shows that B/A values for toluene, using eq. (6), generally decrease with increase of pressure. In this case, some values of B/A are found to be negative. It is because of the value of x which is greater than 1.2, the maximum limit of x in the Tong-Dong method. In fact, in the Tong-Dong method, the B/A values are highly sensitive to the values of x. It has been noticed that even small changes in the values of x give very absurd values of B/A. This is mainly due to

Table 1. Nonlinearity parameter (B/A) of toluene at 303.15 K as a function of pressure along with density and ultrasonic velocity.

P (MPa)	ρ (g cm^{-3})	u (m s^{-1})	B/A Ballou's	B/A Hartmann's	B/A Tong-Dong	B/A Sharma
0.1	0.8580	1258.8	8.833	9.622	−15.874	7.450
10	0.8657	1334.3	8.493	9.345	−66.042	7.490
20	0.8729	1378.7	8.204	9.108	62.940	7.526
30	0.8796	1421.6	7.941	8.894	26.285	7.560
40	0.8858	1461.3	7.712	8.706	18.614	7.590
50	0.8917	1499.9	7.501	8.534	15.235	7.618
60	0.8972	1535.1	7.317	8.384	13.453	7.645
70	0.9025	1567.0	7.158	8.254	12.378	7.671
80	0.9075	1601.0	6.995	8.121	11.572	7.694
90	0.9122	1630.3	6.861	8.011	11.056	7.717
100	0.9168	1659.3	6.732	7.906	10.662	7.739
110	0.9212	1688.0	6.609	7.806	10.356	7.590
120	0.9253	1716.1	6.493	7.711	10.118	7.779
130	0.9294	1742.0	6.389	7.626	9.941	7.798
140	0.9333	1769.2	6.283	7.539	9.790	7.816
150	0.9370	1794.0	6.189	7.463	9.679	7.834
160	0.9407	1816.0	6.108	7.396	9.597	7.852

Table 2. Nonlinearity parameter (B/A) of o-xylene at 303.15 K as a function of pressure along with density and ultrasonic velocity.

P (MPa)	ρ (g cm^{-3})	u (m s^{-1})	B/A Ballou's	B/A Hartmann's	B/A Tong-Dong	B/A Sharma
0.1	0.8714	1329.5	8.526	9.371	22.422	7.437
10	0.8784	1377.4	8.212	9.115	16.125	7.475
20	0.8850	1418.4	7.960	8.909	13.644	7.511
30	0.8912	1459.8	7.720	8.713	12.185	7.543
40	0.8971	1504.1	7.478	8.516	11.207	7.572
50	0.9026	1539.8	7.293	8.364	10.665	7.600
60	0.9079	1584.0	7.076	8.187	10.198	7.623
70	0.9129	1607.3	6.966	8.097	10.006	7.650
80	0.9176	1638.7	6.823	7.980	9.805	7.674
90	0.9222	1667.7	6.696	7.876	9.661	7.696
100	0.9266	1690.6	6.598	7.797	9.569	7.720
110	0.9308	1721.8	6.496	7.692	9.473	7.739
120	0.9349	1748.9	6.361	7.604	9.411	7.759
130	0.9388	1773.2	6.267	7.527	9.368	7.778
140	0.9426	1797.0	6.178	7.454	9.338	7.797
150	0.9463	1820.4	6.092	7.383	9.317	7.815
160	0.9498	1847.6	5.995	7.304	9.305	7.831

Table 3. Nonlinearity parameter (B/A) of aniline at 303.15 K as a function of pressure along with density and ultrasonic velocity.

P (MPa)	ρ (g cm^{-3})	u (m s^{-1})	B/A Ballou's	B/A Hartmann's	B/A Tong-Dong	B/A Sharma
0.1	0.9871	1624.2	6.888	8.034	9.502	7.374
10	0.9917	1653.3	6.758	7.928	9.408	7.403
20	0.9962	1682.1	6.634	7.826	9.337	7.431
30	1.0005	1710.1	6.517	7.731	9.285	7.457
40	1.0047	1737.3	6.407	7.641	9.249	7.482
50	1.0088	1763.9	6.303	7.556	9.225	7.505
60	1.0127	1787.7	6.213	7.482	9.212	7.529
70	1.0166	1814.9	6.112	7.400	9.207	7.550
80	1.0203	1839.3	6.024	7.328	9.209	7.571
90	1.0240	1862.9	5.942	7.261	9.217	7.591
100	1.0275	1885.9	5.863	7.196	9.229	7.611
110	1.0310	1908.1	5.789	7.136	9.245	7.630
120	1.0343	1929.7	5.719	7.079	9.264	7.648
130	1.0376	1950.5	5.652	7.024	9.285	7.666
140	1.0409	1970.5	5.590	6.973	9.309	7.683
150	1.0440	1989.9	5.530	6.925	9.334	7.700
160	1.0471	2008.5	5.475	6.879	9.359	7.717

the factors ($3-2x$) in the numerator and ($6-5x$) in the denominator in the mathematical expression for $J(x)$ given by eq. (8). In fact, good values of $J(x)$ or B/A result when x lies between 1.10 and 1.14. It is clear from Table 2 that B/A values for o-xylene decrease with increase of pressure in this method. Table 3 reveals that in the case of aniline, there is a decrease in B/A values with increase of pressure upto 70 MPa, but after this value of pressure, B/A values show an increasing trend with pressure. Nonlinearity parameter is related with molecular interactions between components. Among the three pure components, aniline molecules are known to be associated through hydrogen bonding in the pure state, which would be affected largely by pressure change. It is evident from eqs. (1) and (2) that Ballou and Hartmann relations depend mainly on the ultrasonic velocity. On increasing the pressure, an increase in ultrasonic velocity has been observed, thereby causing a decrease in B/A values from the two relations. In the expression given by Sharma, factor affecting the nonlinearity parameter is thermal expansion coefficient.

Tables 4, 5 reveal that for both the liquid mixtures, the B/A values calculated by Ballou's empirical relation and Hartmann's expression decrease as the pressure increases, but the value of B/A computed by Sharma method show an opposite trend *i.e.* it increases as the pressure is increased. However, the variation in the B/A values due to this method is very small in comparison with the other methods. B/A values calculated by Tong-Dong method initially decrease with pressure upto a certain pressure, and after that it increases with the increase of pressure for toluene + o-xylene. The value of pressure at which the trend changes, decrease as the mole fraction of toluene decreases in the mixture. The same trend is also observed for toluene + aniline but only when the content of toluene in the mixture is greater than that of aniline. This trend is not present when the content of toluene is less than that of aniline in the mixture. Also, the nonlinearity parameter decreases, in general, with the decrease in concentration of toluene in both the mixtures for all the methods except Tong-Dong. In the Tong-Dong method, this type of trend is observed upto a certain pressure. But, after this pressure, the B/A values tend to increase as the mole fraction of toluene in the mixtures decreases. Variation of ultrasonic velocity with pressure in both the binary mixtures is different. In the binary mixture of toluene + o-xylene, variation of ultrasonic velocity with mole fraction of toluene (x_1) is concave at atmospheric pressure and passes

Table 4. Nonlinearity parameter B/A of x-toluene +($1-x$) o-xylene at different mole fractions at 303.15 K as a function of pressure.

Pressure (MPa)	B/A ($x = 0.8$) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.792	9.588	9.798	7.447
10	8.436	9.298	7.561	7.470
20	8.140	9.056	9.421	7.492
30	7.898	8.858	9.339	7.513
40	7.657	8.662	9.284	7.532
50	7.456	8.497	9.257	7.550
60	7.266	8.342	9.247	7.567
70	7.110	8.215	9.249	7.585
80	6.950	8.084	9.262	7.601
90	6.990	8.117	9.279	7.617
100	6.698	7.878	9.299	7.632
110	6.570	7.774	9.328	7.647
120	6.449	7.675	9.361	7.661
130	6.348	7.593	9.362	7.675
140	6.260	7.521	9.421	7.689
150	6.172	7.449	9.454	7.702
160	6.084	7.376	9.491	7.715

Pressure (MPa)	B/A ($x = 0.6$) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.712	9.523	9.622	7.445
10	8.376	9.249	9.449	7.468
20	8.089	9.015	9.343	7.490
30	7.838	8.809	9.283	7.511
40	7.606	8.620	9.251	7.530
50	7.416	8.465	9.241	7.548
60	7.221	8.305	9.244	7.572
70	7.084	8.194	9.254	7.594
80	6.922	8.061	9.276	7.599
90	6.783	7.948	9.302	7.615
100	6.664	7.851	9.329	7.634
110	6.534	7.774	9.366	7.653
120	6.424	7.654	9.401	7.660
130	6.319	7.569	9.438	7.674
140	6.234	7.499	9.470	7.690
150	6.146	7.428	9.507	7.701
160	6.058	7.356	9.547	7.717

Table 4. *(Contd.)*

Pressure (MPa)	B/A (x = 0.4) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.656	9.478	9.501	7.442
10	8.325	9.207	9.369	7.466
20	8.044	9.978	9.286	7.488
30	7.803	8.781	9.249	7.509
40	7.564	8.586	9.233	7.528
50	7.369	8.426	9.236	7.547
60	7.183	8.274	9.252	7.570
70	7.031	8.150	9.272	7.592
80	6.887	8.033	9.300	7.598
90	6.747	7.918	9.333	7.614
100	6.640	7.831	9.362	7.633
110	6.523	7.735	9.399	7.651
120	6.399	7.634	9.446	7.659
130	6.299	7.552	9.486	7.673
140	6.201	7.473	9.528	7.690
150	6.127	7.412	9.561	7.701
160	6.042	7.343	9.602	7.716

Pressure (MPa)	B/A (x = 0.2) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.588	9.422	9.402	7.439
10	8.269	9.161	9.307	7.463
20	7.999	8.941	9.247	7.486
30	7.755	8.741	9.227	7.507
40	7.519	8.549	9.228	7.526
50	7.329	8.394	9.242	7.544
60	7.156	8.252	9.266	7.568
70	6.992	8.118	9.298	7.590
80	6.847	8.000	9.333	7.596
90	6.721	7.897	9.368	7.612
100	6.623	7.817	9.397	7.631
110	6.495	7.713	9.445	7.650
120	6.378	7.617	9.493	7.658
130	6.279	7.536	9.536	7.672
140	6.195	7.468	9.574	7.688
150	6.107	7.395	9.619	7.700
160	6.019	7.324	9.666	7.715

Table 5. Nonlinearity parameter *B/A* of *x*-toluene + (1–*x*) aniline at different mole fractions at 303.15 K as a function of pressure.

Pressure (MPa)	B/A (x = 0.8) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.471	9.327	9.506	7.430
10	8.196	9.101	9.385	7.454
20	7.942	8.895	9.305	7.477
30	7.711	8.706	9.256	7.498
40	7.490	8.526	9.230	7.518
50	7.310	8.378	9.222	7.537
60	7.132	8.233	9.227	7.556
70	6.990	8.117	9.239	7.574
80	6.839	7.994	9.260	7.590
90	6.719	7.895	9.282	7.607
100	6.588	7.789	9.313	7.623
110	6.474	7.695	9.345	7.638
120	6.363	7.605	9.381	7.653
130	6.279	7.536	9.409	7.668
140	6.173	7.450	9.452	7.682
150	6.097	7.388	9.482	7.696
160	6.012	7.318	9.521	7.710

Pressure (MPa)	B/A (x = 0.6) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	8.121	9.040	9.256	7.428
10	7.865	8.831	9.213	7.443
20	7.632	8.641	9.194	7.461
30	7.447	8.490	9.192	7.483
40	7.261	8.338	9.203	7.506
50	7.095	8.203	9.222	7.524
60	6.943	8.078	9.247	7.548
70	6.803	7.964	9.278	7.566
80	6.678	7.862	9.310	7.579
90	6.555	7.761	9.348	5.596
100	6.430	7.660	9.392	7.614
110	6.322	7.580	9.429	7.632
120	6.230	7.496	9.472	7.644
130	6.147	7.428	9.509	7.659
140	6.069	7.365	9.545	7.675
150	5.985	7.296	9.588	7.689
160	5.909	7.234	9.628	7.704

Table 5. *(Contd.)*

Pressure (MPa)	B/A (x = 0.4) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	7.724	8.717	9.157	7.417
10	7.519	8.549	9.166	7.430
20	7.320	8.386	9.188	7.447
30	7.159	8.255	9.213	7.469
40	6.998	8.124	9.247	7.494
50	6.853	8.005	9.285	7.511
60	6.729	7.903	9.322	7.536
70	6.593	7.793	9.369	7.552
80	6.490	7.708	9.407	7.568
90	6.397	7.633	9.443	7.585
100	6.280	7.537	9.498	7.604
110	6.182	7.457	9.545	7.621
120	6.083	7.376	9.598	7.635
130	6.009	7.316	9.637	7.650
140	5.934	7.254	9.679	7.667
150	5.862	7.195	9.722	7.681
160	5.790	7.137	9.767	7.696

Pressure (MPa)	B/A (x = 0.2) Ballou's	Hartmann's	Tong-Dong	Sharma
0.1	7.858	8.381	9.196	7.405
10	7.146	8.244	9.234	7.418
20	6.985	8.112	9.278	7.434
30	6.842	7.996	9.323	7.457
40	6.722	7.898	9.364	7.482
50	6.606	7.803	9.408	7.500
60	6.475	7.696	9.466	7.524
70	6.368	7.609	9.515	7.540
80	6.276	7.534	9.559	7.558
90	6.188	7.462	9.605	7.574
100	6.088	7.380	9.662	7.594
110	6.005	7.313	9.710	7.611
120	5.920	7.243	9.764	7.626
130	5.853	7.189	9.805	7.641
140	5.790	7.137	9.845	7.658
150	5.720	7.080	9.894	7.673
160	5.655	7.027	9.941	7.688

into a convex curve with rise of pressure. In the binary mixture of toluene + aniline, variation of ultrasonic velocity with x_1, is concave over the whole experimental range. The effect of pressure as observed in the ultrasonic velocity values, can also be seen in *B/A* values calculated from Tong-Dong method in both the binary mixtures.

The interaction study made earlier [27] has also shown a marked effect of pressure on excess isentropic compressibility values k_s^E. The sign of k_s^E changes from positive to negative, and negative to positive with rise in pressure for binary mixtures toluene + o-xylene and toluene + aniline, respectively. Moreover, the maxima of k_s^E curve shifts largely in the toluene-rich region at high pressure in the binary mixture toluene + aniline, while k_s^E for toluene + o-xylene show a symmetrical behaviour. In the present investigation, we have also observed that variation of *B/A* from Tong-Dong method is sharply affected by the toluene content in toluene + aniline mixture.

On the basis of the above discussion, it can be concluded that the variation of computed values of *B/A* from Tong-Dong method resembles with the studies made earlier, and thus confirms the superiority of this method over others.

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